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## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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<b>(21) International Application Number:</b> PCT/US97/20512 <b>(22) International Filing Date:</b> 6 November 1997 (06.11.97) <b>(30) Priority Data:</b> 96870146.6 13 November 1996 (13.11.96) EP <i>(34) Countries for which the regional or international application was filed:</i> BE et al. <b>(71) Applicant (for all designated States except US):</b> THE PROCTER & GAMBLE COMPANY [US/US]; One Procter & Gamble Plaza, Cincinnati, OH 45202 (US). <b>(72) Inventors; and</b> <b>(75) Inventors/Applicants (for US only):</b> PETRI, Marco [IT/IT]; Via Ai Boschi, 70, I-21021 Angera Varese (IT). TRANI, Marina [IT/IT]; Via Gualtiero Serafino, 20, I-00136 Rome (IT). ROMANO, Nicoletta [IT/IT]; Via Calcutta, 25, I-00144 Rome (IT). SEREGO ALLIGHIERI, Giadra [IT/IT]; Via Adelaide Ristori, 32/b, I-00197 Rome (IT). <b>(74) Agents:</b> REED, T., David et al.; The Procter & Gamble Company, 5299 Spring Grove Avenue, Cincinnati, OH 45217 (US).		<b>(81) Designated States:</b> AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GE, GH, HU, ID, IL, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, US, UZ, VN, YU, ZW, ARIPO patent (GH, KE, LS, MW, SD, SZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, ML, MR, NE, SN, TD, TG).  <b>Published</b> <i>With international search report.</i>
<b>(54) Title:</b> DISINFECTING MICROEMULSIONS  <b>(57) Abstract</b>  In its broadest embodiment the present invention relates to the use of a microemulsion comprising a surfactant, an aqueous phase and droplets dispersed in said aqueous phase, said droplets comprising an essential oil or an active thereof, and said droplets having a particle size of less than 100 nanometers, for disinfecting a surface. The present invention further encompasses a microemulsion suitable for disinfecting a surface, comprising a surfactant, an aqueous phase comprising a bleach, and droplets dispersed in said aqueous phase, said droplets comprising an essential oil or an active thereof, and said droplets having a particle size of less than 100 nanometers.		

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## DISINFECTING MICROEMULSIONS

### Technical field

The present invention relates to compositions which can be used to disinfect various surfaces including animate surfaces (e.g., human skin, mouth and the like) and inanimate surfaces including, but not limited to, hard surfaces like walls, tiles, floors, glass, bathroom surfaces, kitchen surfaces, dishes as well as fabrics, clothes, carpets and the like.

### Background of the invention

Antimicrobial/antibacterial compositions include materials which have the ability to disinfect. It is generally recognised that a disinfecting material greatly reduces or even eliminates the microorganisms, e.g., bacteria, existing on a surface. For example compositions based on quaternary ammonium compounds have been extensively described in the art for disinfecting purpose.

Although such disinfecting compositions provide acceptable disinfecting properties they do not encounter good acceptance amongst the consumers who are looking for disinfecting compositions based on safer and less harsh chemicals.

It is thus an object of the present invention to provide compositions which are effective disinfectants on various surfaces, and which are safe to the surfaces treated and the environment.

It has now been found that this can be achieved by formulating an essential oil or an active thereof in the form of a microemulsion having droplets dispersed in an aqueous phase, said droplets comprising said essential oil or an active thereof, and said droplets having a particle size of less than 100 nm. Indeed, it has surprisingly been found that improved disinfecting performance is provided with a microemulsion of the present invention, as compared to a similar composition which is not in the form of a microemulsion as defined in the present invention.

Thus, in its broadest embodiment the present invention encompasses the use of a microemulsion comprising a surfactant, an aqueous phase and droplets dispersed in said aqueous phase, said droplets comprising an essential oil or an active thereof, and said droplets having a particle size of less than 100 nanometers, for disinfecting a surface. The present invention also encompasses a microemulsion suitable for disinfecting a surface comprising a surfactant, an aqueous phase comprising a bleach, preferably a peroxygen bleach, and droplets dispersed in said aqueous phase, said droplets comprising an essential oil or an active thereof, and said droplets having a particle size of less than 100 nanometers.

An advantage of the present invention is that effective disinfecting performance is delivered on a broad range of bacterial strains including Gram positive and Gram negative bacterial strains but also more resistant micro-organisms like fungi, even at high dilution levels, e.g., up to dilution levels of from 1:100 (microemulsion:water).

Another advantage of the present invention is that besides the disinfection properties delivered, good cleaning is also provided as the microemulsions herein comprise at least one surfactant and optionally a solvent.

Also the microemulsions according to the present invention are suitable for disinfecting various surfaces including animate surfaces (e.g. human skin

and/or mouth) as well as inanimate surfaces. Indeed, this technology is particularly suitable in hard-surfaces applications, laundry applications, e.g., in a so-called "soaking mode", "through the wash mode" and/or "pretreatment mode", as well as in carpet applications and the like.

### Background art

WO 96/26262 discloses microemulsion light duty liquid cleaning compositions comprising 1% to 26% by weight of at least one anionic surfactant selected from the group consisting of sulfonate surfactants, alkyl sulfate surfactants and ethoxylated alkyl ether sulfate surfactants and mixtures thereof, 0% to 25% by weight of a zwitterionic surfactant, 0.5% to 29% by weight of a biodegradable compound selected from the group consisting of a mixture of an ethoxylated nonesterified polyhydric alcohol, an ethoxylated fully esterified polyhydric alcohol and an ethoxylated partially esterified polyhydric alcohol, 0.4% to 10% of a water insoluble hydrocarbon, essential oil or a perfume, 1% to 20% of a cosurfactant and the balance being water. No bleaches are disclosed. No disinfecting application is disclosed.

US 5 468 725 discloses an alcohol-free transparent perfume consisting essentially of an alcohol-free perfume base, water and a stable transparent oil-in-water microemulsion fragrance concentrate consisting of water, at least one hydrophobic perfume oil (e.g., lavender oil, geraniol), at least one cationic surfactant and at least one non-ionic surfactant, wherein the transparent microemulsion perfume has a refractive index in the range of from 1.4 up to 1.6 at a temperature in the range of from 20°C up to 30°C. No bleaches are disclosed. Also no disinfection application is disclosed.

WO 96/01305 discloses an aqueous cleaning composition which upon aqueous dilution by a factor of at least two produces a stable microemulsion, said emulsion having a measured dispersed phase particle size of 10-100 nanometers, said composition including water, surfactant (15%-40%), solvent (5%-30%), water insoluble oil (5%-20%), said composition having a

measured dispersed phase particle size of greater than 100 nm prior to dilution. No bleaches are disclosed. No disinfection application is disclosed.

### Summary of the invention

The present invention encompasses a microemulsion suitable for disinfecting a surface, comprising a surfactant, an aqueous phase comprising a bleach, and droplets dispersed in said aqueous phase, said droplets comprising an essential oil or an active thereof, and said droplets having a particle size of less than 100 nanometers.

In its broadest embodiment the present invention encompasses the use of a microemulsion comprising a surfactant, an aqueous phase and droplets dispersed in said aqueous phase, said droplets comprising an essential oil or an active thereof, said droplets having a particle size of less than 100 nanometers, for disinfecting a surface.

### Detailed description of the invention

#### Microemulsions of the present invention:

The present invention encompasses a microemulsion suitable for disinfecting a surface comprising a surfactant, an aqueous phase comprising a bleach, and droplets dispersed in said aqueous phase, said droplets comprising an essential oil or an active thereof, and said droplets having a particle size of less than 100 nanometers.

The microemulsions of the present invention that may also be called "swollen micellar solutions" or "solubilized micellar solutions" have a transparent or translucent appearance, as opposed to an opaque or milk appearance typically associated with emulsions. The microemulsions herein are also physically stable. By "physically stable" it is meant herein that the

microemulsions do not show phase separation upon prolonged storage, i.e., the droplets comprising essential oils/actives remain dispersed in the aqueous phase.

The present invention is based on the finding that improved disinfecting performance is provided when a disinfecting composition comprising a surfactant, a bleach preferably a peroxygen bleach, an essential oil or an active thereof and an aqueous phase is formulated in the form of a microemulsion comprising droplets dispersed in said aqueous phase, said droplets comprising said essential oil or an active thereof, and having a particle size of less than 100 nanometers, as compared to a similar composition which is not in the form of a microemulsion according to the present invention. Indeed, effective disinfecting performance is delivered with the microemulsions according to the present invention with a low total level of disinfecting actives.

By "effective disinfecting performance" it is meant herein that the microemulsions of the present invention allow to significantly reduce the amount of bacteria on an infected surface. Indeed, effective disinfection is obtained on various microorganisms including Gram positive bacteria like *Staphylococcus aureus*, and Gram negative bacteria like *Pseudomonas aeruginosa*, as well as on fungi like *Candida albicans* present on infected surfaces.

The disinfecting performance of a composition may be measured by the bactericidal activity of said composition. A test method suitable to evaluate the bactericidal activity of a composition on a surface is described in European Standard, prEN 1040, CEN/TC 216 N 78, dated November 1995 issued by the European committee for standardisation, Brussels. European Standard, prEN 1040, CEN/TC 216 N 78, specifies a test method and requirements for the minimum bactericidal activity of a disinfecting composition. The test is passed if the bacterial colonies forming units (cfu) are reduced from a  $10^7$  cfu (initial level) to a  $10^2$  cfu (final level after contact with the disinfecting product), i.e., a  $10^5$  reduction of the viability is necessary. The microemulsions according to the present invention pass this test, even if used in highly diluted conditions, e.g. up to a dilution level of 100:1 (water:microemulsion).



Preferably in the microemulsions according to the present invention as is said droplets comprising said essential oil or active thereof, have a particle size of less than 90 nm, preferably less than 80 nm. Dilution upon use of the microemulsions of the present invention does not affect the particle size of said droplets inasmuch that the particle size of said droplets is less than 100 nm, preferably less than 90 nm upon dilution.

A test method suitable to evaluate the size of the droplets comprising said essential oil or an active thereof in the microemulsions according to the present invention is Cryo-transmission electron microscopy (Cryo-TEM). Cryo-TEM samples are prepared in a controlled environment vitrification system (CEVS) which is described in detail in Bellare, J. R.; Davis, H. T.; Scriven, L. E.; Talmon, Y., Controlled environment vitrification system (CEVS): An improved sample preparation technique, J. Electron Microsc. Tech., 1988, 10, 87-111. A 5  $\mu$ l drop of the sample microemulsion is placed on a carbon-coated holey polymer support film mounted on the surface of a standard 200-mesh TEM grid (Ted Pella, Inc., Catalog # 01883). The drop is blotted with filter paper until it is reduced to a thin film (10-200 nm) of the sample spanning the holes (2-8  $\mu$ m) of the support film. The sample is then vitrified by rapidly plunging it through a synchronous shutter at the bottom of the CEVS into liquid ethane at its freezing point. The vitreous specimen is transferred under liquid nitrogen into a Philips CM12<sup>®</sup> microscope for imaging. The temperature of the sample is kept under -170°C throughout the examination.

An essential element of the present invention is an essential oil or an active thereof or mixtures thereof.

Suitable essential oils or actives thereof to be used in the microemulsions herein are those essential oils which exhibit antimicrobial activity and more particularly antibacterial activity. By "actives of essential oils" it is meant herein any ingredient of essential oils that exhibit antimicrobial/antibacterial activity. It is speculated that said essential oils and actives thereof act as proteins denaturing agents. A further advantage of said essential oils and actives hereof is that they impart pleasant odor to the microemulsions

according to the present invention without the need of adding a perfume. Indeed, the microemulsions according to the present invention deliver not only excellent disinfecting performance on infected surfaces but also good scent.

Such essential oils include, but are not limited to, those obtained from thyme, lemongrass, citrus, lemons, oranges, anise, clove, aniseed, cinnamon, geranium, roses, mint, lavender, citronella, eucalyptus, peppermint, camphor, sandalwood and cedar and mixtures thereof.

Actives of essential oils to be used herein include, but are not limited to, thymol (present for example in thyme), eugenol (present for example in cinnamon and clove), menthol (present for example in mint), geraniol (present for example in geranium and rose), verbenone (present for example in vervain), eucalyptol and pinocarvone (present in eucalyptus), cedrol (present for example in cedar), anethol (present for example in anise), carvacrol, hinokitiol, berberine, terpineol, limonene, and mixtures thereof. Preferred actives of essential oils to be used herein are thymol, eugenol, verbenone, eucalyptol, limonene and/or geraniol.

Thymol may be commercially available for example from Aldrich, eugenol may be commercially available for example from Sigma, Systems - Bioindustries (SBI) - Manheimer Inc.

Typically, the microemulsions according to the present invention comprise from 0.005% to 5% by weight of the total microemulsion of said essential oil or active thereof or mixtures thereof, preferably from 0.006% to 3%, more preferably from 0.05% to 1%.

Another essential element of the present invention is a surfactant or a mixture thereof.

A surfactant is needed in order to form the microemulsions according to the present invention because it allows to disperse the oily phase, i.e. droplets comprising the essential oil or an active thereof, in the aqueous phase of the oil-in-water microemulsions of the present invention. In other words, in the absence of any surfactant the present microemulsions would not form

because the dispersed oily phase, i.e. the droplets comprising the essential oil or an active thereof, would tend to quickly grow and separate from the aqueous phase. Thus, the presence of a surfactant or mixtures thereof allows to control the size of the droplets comprising said essential oil or active thereof according to the present invention.

It is understood herein that the surfactant to be used herein or mixtures thereof as well as the levels thereof are chosen, depending on the nature and level of the essential oil or active thereof, so as to form the microemulsions according to the present invention.

Typically, the microemulsions according to the present invention comprise from 0.01% to 40% by weight of the total microemulsion of a surfactant, or mixtures thereof, preferably from 0.05 % to 15% and more preferably from 0.1% to 12 %.

Suitable surfactants to be used in the present invention include any surfactant known to those skilled in the art as being able to form a microemulsion as defined herein, comprising droplets comprising the essential oil or an active thereof, when adding to an aqueous composition comprising said essential oil or an active thereof. Suitable surfactants include nonionic, anionic, cationic, amphoteric and/or zwitterionic surfactants. Said surfactants are also desirable herein as they contribute to the cleaning performance of the present microemulsions.

Preferred surfactants to be used herein are the zwitterionic and/or amphoteric surfactants.

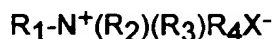
Suitable amphoteric surfactants to be used herein include amine oxides having the following formula  $R_1R_2R_3NO$  wherein each of  $R_1$ ,  $R_2$  and  $R_3$  is independently a saturated substituted or unsubstituted, linear or branched hydrocarbon chains of from 1 to 30 carbon atoms. Preferred amine oxide surfactants to be used according to the present invention are amine oxides having the following formula  $R_1R_2R_3NO$  wherein  $R_1$  is an hydrocarbon chain comprising from 1 to 30 carbon atoms, preferably from 6 to 20, more preferably from 8 to 16, most preferably from 8 to 12, and wherein  $R_2$  and  $R_3$  are independently substituted or unsubstituted, linear or branched

hydrocarbon chains comprising from 1 to 4 carbon atoms, preferably from 1 to 3 carbon atoms, and more preferably are methyl groups. R<sub>1</sub> may be a saturated substituted or unsubstituted linear or branched hydrocarbon chain.

Suitable amine oxides for use herein are for instance natural blend C8-C10 amine oxides as well as C12-C16 amine oxides commercially available from Hoechst.

Amine oxides are preferred herein as they deliver effective cleaning performance and further participate to the disinfecting properties of the microemulsions herein.

Suitable zwitterionic surfactants to be used herein contain both cationic and anionic hydrophilic groups on the same molecule at a relatively wide range of pH's. The typical cationic group is a quaternary ammonium group, although other positively charged groups like phosphonium, imidazolium and sulfonium groups can be used. The typical anionic hydrophilic groups are carboxylates and sulfonates, although other groups like sulfates, phosphonates, and the like can be used. A generic formula for some zwitterionic surfactants to be used herein is

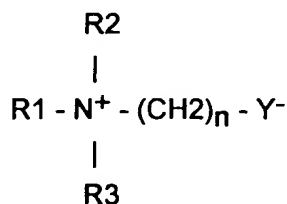


wherein R<sub>1</sub> is a hydrophobic group; R<sub>2</sub> and R<sub>3</sub> are each C<sub>1</sub>-C<sub>4</sub> alkyl, hydroxy alkyl or other substituted alkyl group which can also be joined to form ring structures with the N; R<sub>4</sub> is a moiety joining the cationic nitrogen atom to the hydrophilic group and is typically an alkylene, hydroxy alkylene, or polyalkoxy group containing from 1 to 10 carbon atoms; and X is the hydrophilic group which is preferably a carboxylate or sulfonate group. Preferred hydrophobic groups R<sub>1</sub> are alkyl groups containing from 1 to 24, preferably less than 18, more preferably less than 16 carbon atoms. The hydrophobic group can contain unsaturation and/or substituents and/or linking groups such as aryl groups, amido groups, ester groups and the like. In general, the simple alkyl groups are preferred for cost and stability reasons.

Highly preferred zwitterionic surfactants include betaine and sulphobetaine surfactants, derivatives thereof or mixtures thereof. Said betaine or sulphobetaine surfactants are preferred herein as they help disinfection by increasing the permeability of the bacterial cell wall, thus allowing other active ingredients to enter the cell.

Furthermore, due to the mild action profile of said betaine or sulphobetaine surfactants, they are particularly suitable for the cleaning of delicate surfaces, e.g., delicate laundry or surfaces in contact with food and/or babies. Betaine and sulphobetaine surfactants are also extremely mild to the skin and/or surfaces to be treated.

Suitable betaine and sulphobetaine surfactants to be used herein are the betaine/sulphobetaine and betaine-like detergents wherein the molecule contains both basic and acidic groups which form an inner salt giving the molecule both cationic and anionic hydrophilic groups over a broad range of pH values. Some common examples of these detergents are described in U.S. Pat. Nos. 2,082,275, 2,702,279 and 2,255,082, incorporated herein by reference. Preferred betaine and sulphobetaine surfactants herein are according to the formula

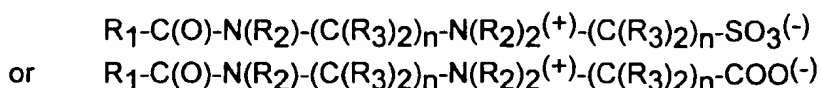


wherein R1 is a hydrocarbon chain containing from 1 to 24 carbon atoms, preferably from 8 to 18, more preferably from 12 to 14, wherein R2 and R3 are hydrocarbon chains containing from 1 to 3 carbon atoms, preferably 1 carbon atom, wherein n is an integer from 1 to 10, preferably from 1 to 6, more preferably is 1, Y is selected from the group consisting of carboxyl and sulfonyl radicals and wherein the sum of R1, R2 and R3 hydrocarbon chains is from 14 to 24 carbon atoms, or mixtures thereof.

Examples of particularly suitable betaine surfactants include C12-C18 alkyl dimethyl betaine such as coconut-betaine and C10-C16 alkyl dimethyl

betaine such as laurylbetaine. Coconutbetaine is commercially available from Seppic under the trade name of Amonyl 265®. Laurylbetaine is commercially available from Albright & Wilson under the trade name Empigen BB/L®.

Other specific zwitterionic surfactants have the generic formulas:



wherein each  $R_1$  is a hydrocarbon, e.g. an alkyl group containing from 8 up to 20, preferably up to 18, more preferably up to 16 carbon atoms, each  $R_2$  is either a hydrogen (when attached to the amido nitrogen), short chain alkyl or substituted alkyl containing from one to 4 carbon atoms, preferably groups selected from the group consisting of methyl, ethyl, propyl, hydroxy substituted ethyl or propyl and mixtures thereof, preferably methyl, each  $R_3$  is selected from the group consisting of hydrogen and hydroxy groups and each  $n$  is a number from 1 to 4, preferably from 2 to 3, more preferably 3, with no more than one hydroxy group in any  $(C(R_3)_2)$  moiety. The  $R_1$  groups can be branched and/or unsaturated. The  $R_2$  groups can also be connected to form ring structures. A surfactant of this type is a C<sub>10</sub>-C<sub>14</sub> fatty acylamidopropylene(hydroxypropylene)sulfobetaine that is available from the Sherex Company under the trade name "Varion CAS sulfobetaine" ®.

In a preferred embodiment of the present invention where the microemulsions herein are particularly suitable for the disinfection of a hard-surface, the surfactant is typically a surfactant system comprising an amine oxide and a betaine or sulphobetaine surfactant, preferably in a weight ratio of amine oxide to betaine or sulphobetaine of 1:1 to 100:1, more preferably of 6:1 to 100:1 and most preferably 10:1 to 50:1. The use of such a surfactant system in the microemulsions herein particularly suitable for disinfecting a hard-surface, provides effective cleaning performance and provides shine on the cleaned surfaces, i.e., the amount of filming/streaking left on the cleaned surface that has been treated with said microemulsions is minimal.

Suitable nonionic surfactants to be used herein are fatty alcohol ethoxylates and/or propoxylates which are commercially available with a variety of fatty alcohol chain lengths and a variety of ethoxylation degrees. Indeed, the HLB values of such alkoxylated nonionic surfactants depend essentially on the chain length of the fatty alcohol, the nature of the alkoxylation and the degree of alkoxylation. Surfactant catalogues are available which list a number of surfactants, including nonionics, together with their respective HLB values.

Particularly suitable for use herein as nonionic surfactants are the hydrophobic nonionic surfactants having an HLB (hydrophilic-lipophilic balance) below 16 and more preferably below 15. Those hydrophobic nonionic surfactants have been found to provide good grease cutting properties.

Preferred nonionic surfactants to be used in the microemulsions according to the present invention are surfactants according to the formula  $\text{RO}-(\text{C}_2\text{H}_4\text{O})_n(\text{C}_3\text{H}_6\text{O})_m\text{H}$ , wherein R is a  $\text{C}_6$  to  $\text{C}_{22}$  alkyl chain or a  $\text{C}_6$  to  $\text{C}_{28}$  alkyl benzene chain, and wherein  $n+m$  is from 0 to 20 and  $n$  is from 0 to 15 and  $m$  is from 0 to 20, preferably  $n+m$  is from 1 to 15 and,  $n$  and  $m$  are from 0.5 to 15, more preferably  $n+m$  is from 1 to 10 and,  $n$  and  $m$  are from 0 to 10. The preferred R chains for use herein are the  $\text{C}_8$  to  $\text{C}_{22}$  alkyl chains. Accordingly, suitable hydrophobic nonionic surfactants for use herein are Dobanol<sup>®</sup> 91-2.5 (HLB= 8.1; R is a mixture of  $\text{C}_9$  and  $\text{C}_{11}$  alkyl chains,  $n$  is 2.5 and  $m$  is 0), or Lutensol<sup>®</sup> TO3 (HLB=8; R is a  $\text{C}_{13}$  alkyl chains,  $n$  is 3 and  $m$  is 0), or Lutensol<sup>®</sup> AO3 (HLB=8; R is a mixture of  $\text{C}_{13}$  and  $\text{C}_{15}$  alkyl chains,  $n$  is 3 and  $m$  is 0), or Tergitol<sup>®</sup> 25L3 (HLB= 7.7; R is in the range of  $\text{C}_{12}$  to  $\text{C}_{15}$  alkyl chain length,  $n$  is 3 and  $m$  is 0), or Dobanol<sup>®</sup> 23-3 (HLB=8.1; R is a mixture of  $\text{C}_{12}$  and  $\text{C}_{13}$  alkyl chains,  $n$  is 3 and  $m$  is 0), or Dobanol<sup>®</sup> 23-2 (HLB=6.2; R is a mixture of  $\text{C}_{12}$  and  $\text{C}_{13}$  alkyl chains,  $n$  is 2 and  $m$  is 0), or Dobanol<sup>®</sup> 45-7 (HLB=11.6; R is a mixture of  $\text{C}_{14}$  and  $\text{C}_{15}$  alkyl chains,  $n$  is 7 and  $m$  is 0) Dobanol<sup>®</sup> 23-6.5 (HLB=11.9; R is a mixture of  $\text{C}_{12}$  and  $\text{C}_{13}$  alkyl chains,  $n$  is 6.5 and  $m$  is 0), or Dobanol<sup>®</sup> 25-7 (HLB=12; R is a mixture of  $\text{C}_{12}$  and  $\text{C}_{15}$  alkyl chains,  $n$  is 7 and  $m$  is 0), or Dobanol<sup>®</sup> 91-5 (HLB=11.6; R is a mixture of  $\text{C}_9$  and  $\text{C}_{11}$  alkyl chains,  $n$  is 5 and  $m$  is 0), or Dobanol<sup>®</sup> 91-6 (HLB=12.5 ; R is a mixture of  $\text{C}_9$  and  $\text{C}_{11}$  alkyl chains,  $n$  is 6 and  $m$  is 0), or Dobanol<sup>®</sup> 91-8 (HLB=13.7 ; R is a mixture of  $\text{C}_9$  and  $\text{C}_{11}$  alkyl chains,  $n$  is 8 and  $m$  is 0), Dobanol<sup>®</sup> 91-10

(HLB=14.2 ; R is a mixture of C<sub>9</sub> to C<sub>11</sub> alkyl chains, n is 10 and m is 0), or mixtures thereof. Preferred herein are Dobanol<sup>®</sup> 91-2.5 , or Lutensol<sup>®</sup> TO3, or Lutensol<sup>®</sup> AO3, or Tergitol<sup>®</sup> 25L3, or Dobanol<sup>®</sup> 23-3, or Dobanol<sup>®</sup> 23-2, or Dobanol<sup>®</sup> 23-10, or mixtures thereof. These Dobanol<sup>®</sup> surfactants are commercially available from SHELL. These Lutensol<sup>®</sup> surfactants are commercially available from BASF and these Tergitol<sup>®</sup> surfactants are commercially available from UNION CARBIDE.

Suitable anionic surfactants to be used herein include water soluble salts or acids of the formula ROSO<sub>3</sub>M wherein R is preferably a C<sub>6</sub>-C<sub>24</sub> hydrocarbyl, preferably an alkyl or hydroxyalkyl having a C<sub>8</sub>-C<sub>20</sub> alkyl component, more preferably a C<sub>8</sub>-C<sub>18</sub> alkyl or hydroxyalkyl, and M is H or a cation, e.g., an alkali metal cation (e.g., sodium, potassium, lithium), or ammonium or substituted ammonium (e.g., methyl-, dimethyl-, and trimethyl ammonium cations and quaternary ammonium cations, such as tetramethyl ammonium and dimethyl piperdinium cations and quaternary ammonium cations derived from alkylamines such as ethylamine, diethylamine, triethylamine, and mixtures thereof, and the like).

Other suitable anionic surfactants to be used herein include alkyl-diphenyl-ether-sulphonates and alkyl-carboxylates. Other anionic surfactants can include salts (including, for example, sodium, potassium, ammonium, and substituted ammonium salts such as mono-, di- and triethanolamine salts) of soap, C<sub>9</sub>-C<sub>20</sub> linear alkylbenzenesulfonates, C<sub>8</sub>-C<sub>22</sub> primary or secondary alkanesulfonates, C<sub>8</sub>-C<sub>24</sub> olefinsulfonates, sulfonated polycarboxylic acids prepared by sulfonation of the pyrolyzed product of alkaline earth metal citrates, e.g., as described in British patent specification No. 1,082,179, C<sub>8</sub>-C<sub>24</sub> alkylpolyglycolethersulfates (containing up to 10 moles of ethylene oxide); alkyl ester sulfonates such as C<sub>14</sub>-C<sub>16</sub> methyl ester sulfonates; acyl glycerol sulfonates, fatty oleyl glycerol sulfates, alkyl phenol ethylene oxide ether sulfates, paraffin sulfonates, alkyl phosphates, isethionates such as the acyl isethionates, N-acyl taurates, alkyl succinamates and sulfosuccinates, monoesters of sulfosuccinate (especially saturated and unsaturated C<sub>12</sub>-C<sub>18</sub> monoesters) diesters of sulfosuccinate (especially saturated and unsaturated C<sub>6</sub>-C<sub>14</sub> diesters), acyl sarcosinates, sulfates of alkylpolysaccharides such as the sulfates of alkylpolyglucoside (the nonionic nonsulfated compounds being described below), branched primary alkyl



sulfates, alkyl polyethoxy carboxylates such as those of the formula  $\text{RO}(\text{CH}_2\text{CH}_2\text{O})_k\text{CH}_2\text{COO-M}^+$  wherein R is a  $\text{C}_8\text{-C}_{22}$  alkyl, k is an integer from 0 to 10, and M is a soluble salt-forming cation. Resin acids and hydrogenated resin acids are also suitable, such as rosin, hydrogenated rosin, and resin acids and hydrogenated resin acids present in or derived from tall oil. Further examples are given in "Surface Active Agents and Detergents" (Vol. I and II by Schwartz, Perry and Berch). A variety of such surfactants are also generally disclosed in U.S. Patent 3,929,678, issued December 30, 1975 to Laughlin, et al. at Column 23, line 58 through Column 29, line 23 (herein incorporated by reference).

Preferred anionic surfactants for use in the microemulsions herein are the alkyl benzene sulfonates, alkyl sulfates, alkyl alkoxylated sulfates, paraffin sulfonates and mixtures thereof.

The aqueous phase of the microemulsions of the present invention comprises at least water and a bleach. Said aqueous phase may further comprise any other water-miscible ingredient desired in the microemulsions herein that has a higher affinity toward said aqueous phase than towards the essential oil/active-containing droplets dispersed therein.

Typically, the microemulsions according to the present invention comprise from 60% to 99.5% by weight of the total microemulsion of water, preferably from 80% to 99% and more preferably from 85% to 98%.

The aqueous phase of the microemulsions of the present invention comprises a bleach or a mixture thereof, as an essential ingredient.

Any bleach known to those skilled in the art may be suitable to be used herein including any chlorine bleach as well as any peroxygen bleach.

Suitable chlorine bleaches to be used herein include any compound capable of releasing chlorine when said compound is in contact with water. Suitable chlorine bleaches include alkali metal dichloroisocyanurates as well as alkali metal hypohalites like hypochlorite and/or hypobromite. Preferred chlorine bleaches are alkali metal hypochlorites. Various forms of alkali metal

hypochlorite are commercially available like for instance sodium hypochlorite.

Preferred bleaches for use herein are peroxygen bleaches, more particularly hydrogen peroxide, or a water soluble source thereof, or mixtures thereof. Hydrogen peroxide is most preferred to be used in the microemulsions according to the present invention.

Peroxygen bleaches like hydrogen peroxide are preferred herein as they are generally perceived to be environmentally acceptable. For example the decomposition products of hydrogen peroxide are oxygen and water. Also, it is believed that the presence of said peroxygen bleach, especially hydrogen peroxide, in the microemulsions of the present invention contribute to the disinfection properties of said microemulsions.

As used herein a hydrogen peroxide source refers to any compound which produces perhydroxyl ions when said compound is in contact with water. Suitable water-soluble sources of hydrogen peroxide for use herein include percarbonates, persilicate, persulphate such as monopersulfate, perborates, peroxyacids such as diperoxydodecandioic acid (DPDA), magnesium perphthalic acid, dialkylperoxides, diacylperoxides, preformed percarboxylic acids, organic and inorganic peroxides and/or hydroperoxides and mixtures thereof.

Typically, the microemulsions herein comprise from 0.001% to 15% by weight of the total microemulsion of said bleach or mixtures thereof, preferably from 0.1% to 10%, and more preferably from 0.2% to 5%.

The aqueous microemulsions according to the present invention have a pH as is of from 1 to 12, preferably from 3 to 10, and more preferably from 3 to 9. The pH of the microemulsions can be adjusted by using alkalinising agents or acidifying agents. Examples of alkalinising agents are alkali metal hydroxides, such as potassium and/or sodium hydroxide, or alkali metal oxides such as sodium and/or potassium oxide. Examples of acidifying agents are organic or inorganic acids such as sulfuric acid.

The microemulsions of the present invention may comprise as a preferred optional ingredient, a hydroxylated solvent or a mixture thereof.

Such hydroxylated solvents are suitable herein because they assist/promote the formation of the microemulsions of the present invention on top of the surfactants herein, and thus further contribute to control the size of the droplets comprising an essential oil or an active thereof, and being dispersed in the aqueous phase of the microemulsions of the present invention. Such hydroxylated solvents will at least partially be present in the oily phase of the oil-in-water microemulsions of the present invention, i.e., in the droplets comprising the essential oils/actives.

By "hydroxylated solvent" it is meant herein any hydrocarbon including aliphatic saturated or unsaturated hydrocarbons or aromatic hydrocarbons comprising at least one hydroxyl group (OH).

Suitable hydroxylated solvents include glycol ethers and/or derivatives thereof, polyols, alkoxylated aliphatic or aromatic alcohols, aliphatic or aromatic alcohols, glycols or mixtures thereof.

Suitable glycol ethers and/or derivatives thereof to be used herein include monoglycol ethers and/or derivatives thereof, di-, tri- and poly-glycol ethers and/or derivatives thereof and mixtures thereof.

Suitable monoglycol ethers and derivatives thereof to be used herein include propylene glycol butyl ether, and water-soluble CELLOSOLVE® solvents or mixtures thereof. Preferred Cellosolve® solvents include 2-(Hexyloxy)ethanol (i.e., 2-hexyl Cellosolve®), ethylene glycol ethyl ether (i.e., 2-ethyl Cellosolve®), ethylene glycol butyl ether (i.e., 2-butyl Cellosolve®) or mixtures thereof.

Suitable polyglycol ethers and derivatives thereof to be used herein include n-butoxypropoxypropanol (n-BPP), butyl triglycol ether (BTGE), butyl diglycol ether (BDGE), diethylene glycol butyl ether, water-soluble CARBITOL® solvents or mixtures thereof.

Preferred water-soluble CARBITOL® solvents are compounds of the 2-(2-alkoxyethoxy)ethanol class, 2-(2-alkoxyethoxy)propanol class and/or 2-(2-

alkoxyethoxy)butanol class wherein the alkoxy group is derived from ethyl, propyl, butyl and tert-butyl. A preferred water-soluble carbitol is 2-(2-butoxyethoxy)ethanol also known as butyl carbitol®.

Suitable polyols to be used herein are aliphatic linear or branched saturated or unsaturated hydrocarbons having from 2 to 12 carbon atoms, preferably 4 to 10, and comprising at least 2 hydroxyl groups, preferably from 2 to 4. Suitable polyols herein are diols such as 2-ethyl-1,3-hexanediol, 2,2,4-trimethyl-1,3-pentanediol, methyl-2,4 pentanediol, 1,6-hexanediol or mixture thereof.

Suitable alkoxyated aliphatic or aromatic alcohols to be used herein are according to the formula  $R(A)_n-OH$  wherein R is a linear or branched saturated or unsaturated hydrocarbon chain having from 1 to 20 carbon atoms, preferably from 2 to 15 and more preferably from 2 to 10, or alkyl substituted or non-alkyl substituted aryl group of from 1 to 20 carbon atoms, preferably from 2 to 15 and more preferably from 2 to 10, wherein A is an alkoxy group preferably butoxy, propoxy and/or ethoxy, and n is an integer of from 1 to 5, preferably 1 to 2. Suitable alkoxyated alcohols to be used herein are 1-methoxy-11-dodecanol methoxy propanol, ethoxy propanol and/or propoxy propanol.

Suitable aliphatic or aromatic alcohols to be used herein are according to the formula  $R-OH$  wherein R is a linear or branched saturated or unsaturated hydrocarbon chain having from 1 to 20 carbon atoms, preferably from 2 to 15 and more preferably from 2 to 10, or alkyl substituted or non-alkyl substituted aryl group of from 1 to 20 carbon atoms, preferably from 2 to 15 and more preferably from 2 to 10. Suitable aliphatic alcohols to be used herein include linear alcohols like decanol, ethanol and/or propanol. Suitable aromatic alcohol to be used herein is benzyl alcohol.

Suitable glycols to be used herein are according to the formula  $HO-(CH_2)_n-OH$  wherein n is an integer of 2 to 12. Suitable glycols to be used herein are dodecaneglycol, 1,2-hexanediol and/or propanediol.

Preferred hydroxylated solvents for use herein are ethylene glycol butyl ether, propylene glycol butyl ether, diethylene glycol butyl ether, benzyl alcohol, 2-propanol, ethylene glycol ethyl ether or mixtures thereof.

The hydroxylated solvents may typically be present in the microemulsions of the present invention up to a level of 15% by weight, preferably from 0.2% to 12% by weight and more preferably from 0.5% to 10% by weight of the total microemulsion.

The microemulsions of the present invention may comprise as an optional ingredient, other solvents including terpene or mixtures thereof.

Suitable terpenes to be used herein are mono-and bicyclic terpenes, especially those of the hydrocarbon class, which include the terpinenes, terpinolenes and pinenes and mixtures thereof. Highly preferred materials of this type are dipentene, alpha-pinene and/or beta-pinene. For example, pinene is commercially available from SCM Glidco (Jacksonville) under the name Alpha Pinene P&F®.

Terpenes solvents are suitable herein as they contribute to the cleaning performance of the disinfecting microemulsions of the present invention.

Typically, other solvents apart the hydroxylated ones as defined herein above can be present in the microemulsions of the present invention up to a level of 5% by weight of the total microemulsion, preferably from 0.02% to 3% by weight and more preferably from 0.05% to 1.5%.

The microemulsions herein may further comprise a variety of other optional ingredients such as other antimicrobial/antibacterial compounds, chelating agents, radical scavengers, thickeners, builders, buffers, stabilisers, bleach activators, soil suspenders, dye transfer agents, brighteners, anti dusting agents, enzymes, dispersant, dye transfer inhibitors, pigments, perfumes, and dyes. Depending on their respective hydrophilic/hydrophobic character these optional ingredients are present in the aqueous phase and/or in the droplets as defined herein of the microemulsions of the present invention.

The microemulsions of the present invention may comprise as an optional ingredient another antimicrobial/antibacterial compound, or a mixture thereof.

Suitable antimicrobial/antibacterial compounds for use herein include paraben, glutaraldehyde or mixtures thereof.

Typically, the microemulsions of the present invention comprises up to 5% by weight of the total microemulsion of another antibacterial/antimicrobial compound or mixtures thereof, preferably up to 1%.

Suitable radical scavengers for use herein include the well-known substituted mono and di hydroxy benzenes and derivatives thereof, alkyl- and aryl carboxylates and mixtures thereof. Preferred radical scavengers for use herein include di-tert-butyl hydroxy toluene (BHT), p-hydroxy-toluene, hydroquinone (HQ), di-tert-butyl hydroquinone (DTBHQ), mono-tert-butyl hydroquinone (MTBHQ), tert-butyl-hydroxy anisole, p-hydroxy-anisol, benzoic acid, 2,5-dihydroxy benzoic acid, 2,5-dihydroxyterephthalic acid, toluic acid, catechol, t-butyl catechol, 4-allyl-catechol, 4-acetyl catechol, 2-methoxy-phenol, 2-ethoxy-phenol, 2-methoxy-4-(2-propenyl)phenol, 3,4-dihydroxy benzaldehyde, 2,3-dihydroxy benzaldehyde, benzylamine, 1,1,3-tris(2-methyl-4-hydroxy-5-t-butylphenyl) butane, tert-butyl-hydroxy-aniline, p-hydroxy aniline as well as n-propyl-gallate. Highly preferred for use herein is di-tert-butyl hydroxy toluene, which is for example commercially available from SHELL under the trade name IONOL CP®. These radical scavengers contribute to the stability of the peroxygen bleach-containing microemulsions herein.

Typically, the radical scavenger, or a mixture thereof, is present in the microemulsions of the present invention up to a level of 5% by weight of the total microemulsion, preferably from 0.002% to 3% by weight and more preferably from 0.002% to 1.5%.

Suitable chelating agents to be used herein may be any chelating agent known to those skilled in the art such as the ones selected from the group comprising phosphonate chelating agents, amino carboxylate chelating agents or other carboxylate chelating agents, or polyfunctionally-substituted

aromatic chelating agents or mixtures thereof. It has now been found that the addition of a chelating agent in combination with a surfactant on top of an essential oil and/or an active thereof, in the microemulsions of the present invention further improves the disinfecting properties of said microemulsion.

Such phosphonate chelating agents may include etidronic acid (1-hydroxyethylidene-bisphosphonic acid or HEDP) as well as amino phosphonate compounds, including amino alkylene poly (alkylene phosphonate), alkali metal ethane 1-hydroxy diphosphonates, nitrilo trimethylene phosphonates, ethylene diamine tetra methylene phosphonates, and diethylene triamine penta methylene phosphonates. The phosphonate compounds may be present either in their acid form or as salts of different cations on some or all of their acid functionalities. Preferred phosphonate chelating agents to be used herein are diethylene triamine penta methylene phosphonates. Such phosphonate chelating agents are commercially available from Monsanto under the trade name DEQUEST®.

Polyfunctionally-substituted aromatic chelating agents may also be useful in the compositions herein. See U.S. patent 3,812,044, issued May 21, 1974, to Connor et al. Preferred compounds of this type in acid form are dihydroxydisulfobenzenes such as 1,2-dihydroxy -3,5-disulfobenzene.

A preferred biodegradable chelating agent for use herein is ethylene diamine N,N'- disuccinic acid, or alkali metal, or alkaline earth, ammonium or substitutes ammonium salts thereof or mixtures thereof. Ethylenediamine N,N'- disuccinic acids, especially the (S,S) isomer have been extensively described in US patent 4, 704, 233, November 3, 1987 to Hartman and Perkins. Ethylenediamine N,N'- disuccinic acid is, for instance, commercially available under the tradename ssEDDS® from Palmer Research Laboratories.

Suitable amino carboxylate chelating agents useful herein include ethylene diamine tetra acetate, diethylene triamine pentaacetate, diethylene triamine pentacetate (DTPA), N-hydroxyethylethylenediamine triacetate, nitrilotriacetate, ethylenediamine tetrapropionate, triethylenetetraaminehexaacetate, ethanoldiglycine, propylene diamine tetracetic acid (PDTA) and

methyl glycine di-acetic acid (MGDA), both in their acid form, or in their alkali metal, ammonium, and substituted ammonium salt forms. Particularly suitable to be used herein are diethylene triamine penta acetic acid (DTPA), propylene diamine tetracetic acid (PDTA) which is, for instance, commercially available from BASF under the trade name Trilon FS® and methyl glycine di-acetic acid (MGDA).

Further carboxylate chelating agents to be used herein includes malonic acid, salicylic acid, glycine, aspartic acid, glutamic acid, dipicolinic acid and derivatives thereof, or mixtures thereof.

Typically, the chelating agent, or a mixture thereof, is present in the microemulsions of the present invention at a level of from 0.001% to 5% by weight of the total microemulsion, preferably from 0.002% to 3% by weight and more preferably from 0.002% to 1.5%.

The microemulsions according to the present invention formulated in their liquid form may further comprise as an optional ingredient, a shear thinning polymeric thickener or a mixture thereof.

Such shear thinning polymeric thickeners are suitable herein as they perform a dual function when they are incorporated in the microemulsion according to the present invention, said function being not only to prevent or diminish inhalation by the user of bleach mist/fog when the microemulsion of the present invention is sprayed onto the surface to be disinfected, but also to provide increased contact time of the microemulsion on vertical surfaces, thereby reducing the risk of microemulsion dripping.

Suitable shear thinning polymeric thickeners to be used herein include synthetic and natural occurring polymers. Suitable shear thinning polymeric thickeners for used herein include polyurethane polymers, polyacrylamide polymers, polycarboxylate polymers such as polyacrylic acids and sodium salts thereof, xanthan gum or derivatives thereof, alginate or a derivative thereof, polysaccharide polymers such as substituted cellulose materials like ethoxylated cellulose, carboxymethylcellulose, hydroxymethylcellulose, hydroxypropyl cellulose, hydroxyethyl cellulose and mixtures thereof.



Preferred shear thinning polymeric thickeners for use in the microemulsions of the invention are xanthan gum or derivatives thereof sold by the Kelco Division of Merck under the tradenames KELTROL®, KELZAN AR®, KELZAN D35®, KELZAN S®, KELZAN XZ® and the like.

Xanthan gum is an extra cellular polysaccharide of xanthomonas campestris. Xanthan gum is made by fermentation based on corn sugar or other corn sweetener by-products. Xanthan gum comprises a poly beta-(1→4)-D-Glucopyranosyl backbone chain, similar to that found in cellulose. Aqueous dispersions of xanthan gum and its derivatives exhibit remarkable rheological properties. Xanthan gum exhibits high pseudoplasticity, i.e., over a wide range of concentrations, rapid shear thinning occurs that is generally understood to be instantaneously reversible. Preferred xanthan materials include crosslinked xanthan materials. Xanthan polymers can be crosslinked with a variety of known covalent reacting crosslinking agents reactive with the hydroxyl functionality of large polysaccharide molecules and can also be crosslinked using divalent, trivalent or polyvalent metal ions. Such crosslinked xanthan gels are disclosed in United States Patent No. 4,782,901, which patent is incorporated by reference herein. Suitable crosslinking agents for xanthan materials include metal cations such as Al<sup>+3</sup>, Fe<sup>+3</sup>, Sb<sup>+3</sup>, Zr<sup>+4</sup> and other transition metals, etc. Known organic crosslinking agents can also be used. The preferred crosslinked xanthan agent of the invention is KELZAN AR®, a product of Kelco, a division of Merck Incorporated.

The polycarboxylate polymers for use herein preferably have a molecular weight of from 500,000 to 4,500,000, preferably from 1,000,000 to 4,000,000. Most preferred polymers for use herein contain from 0.5% to 4% by weight of a cross-linking agent, wherein the cross-linking agent tends to interconnect linear strands of the polymers to form the resulting cross-linked products. Suitable cross-linking agents include the polyalkenyl polyethers. Polycarboxylate polymers include the polyacrylate polymers. Others monomers besides acrylic acid can be used to form these polymers including such monomers as maleic anhydride which acts as a source of additional carboxylic groups. The molecular weight per carboxylate group of monomers containing a carboxylate group typically varies from 25 to 200, preferably from 50 to 150, more preferably from 75 to 125. Further other

monomers may be present in the monomeric mixture, if desired, such as ethylene and propylene which act as diluents.

Preferred polycarboxylate polymers for use herein are the polyacrylate polymers. Commercially available polymers of the polyacrylate type include those sold under the trade names Carbopol®, Acrysol® ICS-1, Polygel®, and Sokalan®. Most preferred polyacrylate polymers are the copolymer of acrylic acid and alkyl (C<sub>5</sub>-C<sub>10</sub>) acrylate, commercially available under the tradename Carbopol® 1623, Carbopol® 695 from BF Goodrich, and copolymer of acrylic acid and maleic anhydride, commercially available under the tradename Polygel® DB from 3V Chemical company. Mixtures of any of the polycarboxylate polymers, herein before described, may also be used.

The microemulsions according to the present invention may comprise up to 10% by weight of the total microemulsion of a shear thinning polymeric thickener, or mixtures thereof, preferably from 0.005% to 5% by weight, more preferably from 0.01% to 2% and most preferably from 0.01% to 1%.

No particular mixing order is required to form the oil-in-water microemulsions of the present invention. The microemulsions of the present invention are easily prepared simply by combining all the ingredients in a suitable vessel or container. The order of mixing the ingredients is not particularly important and generally the various ingredients can be added sequentially or all at once. It is not necessary to use elevated temperatures in the formation step and room temperature is sufficient. In a preferred process of making the microemulsions of the present invention an aqueous phase and an oily phase are prepared independently before being mixed together. For example an aqueous phase is prepared by mixing together at least water, a bleach, a surfactant and optional hydrophilic ingredients like chelating agents, buffers and the like, said phase being adjusted to the desired pH. An oily phase is prepared that comprises at least an essential oil/active thereof and optional ingredients like hydroxylated solvents. Then these two phases are mixed together to form the microemulsions of the present invention.

The use of a microemulsion comprising droplets of essential oil/actives for disinfecting a surface

In its broadest embodiment the present invention encompasses the use of a microemulsion comprising a surfactant, an aqueous phase and droplets dispersed in said aqueous phase, said droplets comprising an essential oil or an active thereof, said droplets having a particle size of less than 100 nanometers, for disinfecting a surface.

The aqueous phase of the microemulsions used according to the present invention comprises at least water. All the ingredients of said microemulsions used to disinfect according to the present invention, i.e., essential oils or actives thereof, surfactants and optional ingredients as well as the levels thereof are as defined herein above.

By "surfaces" it is meant herein any animate surface, e.g., human skin, mouth, teeth as well as any inanimate surface. In a preferred embodiment of the present invention the surfaces to be disinfected with a microemulsion as defined herein, are inanimate surfaces.

These inanimate surfaces include, but are not limited to, hard-surfaces typically found in houses like kitchens, bathrooms, or in car interiors, e.g., tiles, walls, floors, chrome, glass, smooth vinyl, any plastic, plastified wood, table top, sinks, cooker tops, dishes, sanitary fittings such as sinks, showers, shower curtains, wash basins, WCs and the like, as well as fabrics including clothes, curtains, drapes, bed linens, bath linens, table cloths, sleeping bags, tents, upholstered furniture and the like, and carpets. Inanimate surfaces also include household appliances including, but not limited to, refrigerators, freezers, washing machines, automatic dryers, ovens, microwave ovens, dishwashers and so on.

Thus, the present invention also encompasses a process of disinfecting a fabric, as the inanimate surface. In such a process a microemulsion, as defined herein, is contacted with the fabrics to be disinfected. This can be done either in a so-called "pretreatment mode", where a microemulsion, as defined herein, is applied neat onto said fabrics before the fabrics are rinsed, or washed then rinsed, or in a "soaking mode" where a microemulsion, as defined herein, is first diluted in an aqueous bath and the

fabrics are immersed and soaked in the bath, before they are rinsed, or in a "through the wash mode", where a microemulsion, as defined herein, is added on top of a wash liquor formed by dissolution or dispersion of a typical laundry detergent.

In the pretreatment mode, it has been found that it is highly preferred that the fabrics be rinsed after they have been contacted with a microemulsion, as defined herein, before said microemulsion has completely dried off, especially in the embodiment herein wherein the microemulsion used comprises a bleach like a peroxygen bleach. Indeed, it has been found that water evaporation contributes to increase the concentration of free radicals onto the surface of the fabrics and, consequently, the rate of chain reaction. Indeed, free radicals typically result from the decomposition of bleach that may be catalysed due to the presence of metal ions on the surface of a fabric and/or to the exposure of the fabrics to UV radiation from sunlight. It is also speculated that an auto-oxidation reaction occurs upon evaporation of water when such microemulsions containing a bleach are left to dry onto the fabrics. For example said reaction of auto-oxidation generates peroxy-radicals which may contribute to the degradation of cellulose. Thus, not leaving such a bleach-containing microemulsions to dry onto the fabric, in a process of pretreating fabrics, contributes to reduce the tensile strength loss when pretreating fabrics with such products.

In the pretreatment mode, the process comprises the steps of applying a microemulsion, as defined herein, neat onto said fabrics, or at least infected portions thereof (i.e., directly applying said liquid microemulsion, as defined herein onto said fabrics without undergoing any dilution), and subsequently rinsing, or washing then rinsing said fabrics. In this mode, the neat microemulsion can optionally be left to act onto said fabrics for a period of time ranging from 1 min. to 1 hour, preferably from 1 minute to 30 minutes, before the fabrics are rinsed, or washed then rinsed, provided that in the embodiment of the present invention wherein said microemulsion comprises a peroxygen bleach it is not left to dry onto said fabrics. For particularly tough stains, it may be appropriate to further rub or brush said fabrics by means of a sponge or a brush, or by rubbing two pieces of fabrics against each other.

In another mode, generally referred to as "soaking", the process comprises the steps of diluting a microemulsion as defined herein, in an aqueous bath so as to form a diluted composition. The dilution level of said microemulsion, in an aqueous bath is typically up to 1:85, preferably up to 1:50 and more preferably about 1:25 (microemulsion:water). The fabrics are then contacted with the aqueous bath comprising the microemulsion, and the fabrics are finally rinsed, or washed then rinsed. Preferably in that embodiment, the fabrics are immersed in the aqueous bath comprising the microemulsion, and also preferably, the fabrics are left to soak therein for a period of time ranging from 1 minute to 48 hours, and preferably from 3 minutes to 24 hours.

In yet another mode which can be considered as a sub-embodiment of "soaking", generally referred to as "through the wash mode", the microemulsion, as defined herein, is used as a so-called laundry additive. And in that embodiment the aqueous bath is formed by dissolving or dispersing a conventional laundry detergent in water. The microemulsion is contacted with the aqueous bath, and the fabrics are then contacted with the aqueous bath containing the microemulsion. Finally, the fabrics are rinsed.

In another embodiment the present invention also encompasses a process of disinfecting a hard-surface, as the inanimate surface. In such a process a microemulsion, as defined herein, is contacted with the hard-surfaces to be disinfected. Thus, the present invention also encompasses a process of disinfecting a hard-surface with a microemulsion, as defined herein, wherein said process comprises the step of applying said microemulsion to said hard-surface, preferably only infected portions thereof, and optionally rinsing said hard-surface.

In the process of disinfecting hard-surfaces according to the present invention the microemulsion, as defined herein, may be applied to the surface to be disinfected in its neat form or in its diluted form typically up to 200 times their weight of water, preferably into 80 to 2 times their weight of water, and more preferably 60 to 2 times.

In the preferred embodiment of the process of the present invention wherein said liquid microemulsion is applied to a hard-surface to be disinfected in its diluted form, it is not necessary to rinse the surface after the microemulsion has been applied, indeed no visible residues are left onto the surface.

#### Packaging form of the microemulsions

The microemulsions herein may be packaged in a variety of suitable detergent packaging known to those skilled in the art. The microemulsions herein may desirably be packaged in manually operated spray dispensing containers, which are usually made of synthetic organic polymeric plastic materials. Accordingly, the present invention also encompasses microemulsions as described herein before packaged in a spray dispenser, preferably in a trigger spray dispenser or in a pump spray dispenser.

For example, said spray-type dispensers allow to uniformly apply to a relatively large area of a surface to be disinfected, the microemulsions of the present invention, thereby contributing to disinfecting properties of said microemulsions. Such spray-type dispensers are particularly suitable to treat vertical surfaces.

Suitable spray-type dispensers to be used according to the present invention include manually operated foam trigger-type dispensers sold for example by Specialty Packaging Products, Inc. or Continental Sprayers, Inc. These types of dispensers are disclosed, for instance, in US-4,701,311 to Dunnining et al. and US-4,646,973 and US-4,538,745 both to Focarracci. Particularly preferred to be used herein are spray-type dispensers such as T 8500® commercially available from Continental Sprayers International, T8900® commercially available from Continental Sprayers Int., or T 8100® commercially available from Canyon, Northern Ireland. In such a dispenser the microemulsion is divided in fine liquid droplets resulting in a spray that is directed onto the surface to be treated. Indeed, in such a spray-type dispenser the microemulsion contained in the body of said dispenser is directed through the spray-type dispenser head via energy communicated to a pumping mechanism by the user as said user activates said pumping

mechanism. More particularly, in said spray-type dispenser head the microemulsion is forced against an obstacle, e.g. a grid or a cone or the like, thereby providing shocks to help atomise the microemulsion, i.e. to help the formation of the spray form of the microemulsion.

The microemulsions of the present invention may also be executed in the form of wipes. By "wipes" it is meant herein disposable towels incorporating a microemulsion according to the present invention. Preferably said wipes are packaged in a plastic box. Accordingly, the present invention also encompasses wipes, e.g., disposable paper towels, impregnated/wetted with a microemulsion as described herein before. The advantage of this execution is a faster usage of a disinfecting microemulsion by the user, this even outside the house, i.e. there is no need to pour the liquid microemulsions according to the present invention on the surfaces to be disinfected and to dry it out with a cloth. In other words, wipes allow disinfecting of surfaces in one step.

The present invention will be further illustrated by the following examples.

### Examples

The following microemulsions were made by mixing the listed ingredients in the listed proportions (weight % unless otherwise specified).

<b>Microemulsions</b> (weight %)	<b>I</b>	<b>II</b>	<b>III</b>	<b>IV</b>	<b>V</b>	<b>VI</b>
Hydrogen peroxide	3.0	3.0	6.8	3.0	1.0	3.0
Betaine*	0.1	0.1	1.5	0.1	0.05	0.2
C10 amine oxide	1.8	1.8	3.0	1.8	0.9	3.0
Geraniol	0.3	0.3	0.3	0.3	—	0.2
Thymol	—	—	—	—	0.05	0.1
Eugenol	—	—	0.1	—	—	—
Eucalyptol	0.1	0.1	—	0.1	0.1	—

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Butyl carbitol®	2.0	2.0	--	2.0	1.5	1.0
Dobanol 91-10®	0.5	1.5	1.6	1.5	--	1.2
Dobanol 23-3®	--	0.6	1.1	0.6	--	--
Benzyl alcohol	2.0	2.0	--	2.0	0.1	--
Limonene	0.2	0.2	--	0.2	0.1	--
Isopropanol	--	--	--	--	1.0	1.0

Water and minors -----up to 100%-----  
H<sub>2</sub>SO<sub>4</sub> up to pH 4

Betaine \* is either coconut betaine commercially available from Seppic under the trade name Amonyl 265® or laurylbetaine commercially available from Albright & Wilson under the trade name Empigen BB/L® or mixtures thereof.

Butyl carbitol® is diethylene glycol butyl ether

Dobanol 91-10® is nonionic surfactant having an aliphatic chain of C9-C11 and an ethoxylation of 10 available from Shell

Dobanol 23-3® is nonionic surfactant having an aliphatic chain of C12-C13 and an ethoxylation of 3 available from Shell.

Microemulsions (weight %)	VII	VIII	IX	X	XI	XII
Hydrogen peroxide	2.0	2.0	3.0	1.0	1.0	1.0
Betaine*	1.5	1.0	1.0	1.0	0.2	0.1
Lauryl amine oxide	1.0	1.0	3.0	3.0	3.0	2.0
Thymol	--	0.1	--	--	--	--
Geraniol	--	--	0.05	0.1	--	--
Eucalyptol	--	--	0.05	--	--	--
Ethyl paraben	--	--	--	--	0.4	0.4
Eugenol	--	--	--	--	--	0.2
Dobanol 91-10®	0.5	0.5	0.3	0.3	0.8	0.1
HEDP	0.1	--	0.1	0.05	0.2	0.3
ATMP	--	0.1	--	--	--	--
BHT	0.1	0.1	0.05	0.05	0.08	0.08
Tetraborate	0.5	0.5	0.7	0.7	1.0	1.0



Water and minors \_\_\_\_\_ up to 100% \_\_\_\_\_  
NaOH up to pH 8.5

HEDP is etidronic acid.

ATMP is nitrilotris(methylene)triphosphonic acid.

BHT is tert-butyl hydroxy toluene.

Tetraborate is sodium tetraborate decahydrate.

These microemulsions are according to the present invention, i.e. that they comprise droplets comprising essential oils/actives, said droplet having a particle size of less than 100 nm, when the microemulsions are both in their neat form or diluted form.

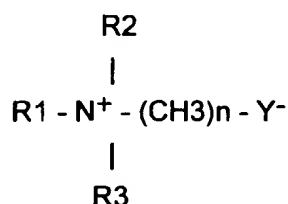
These microemulsions passed the prEN 1040 test of the European committee of standardisation. Indeed, these microemulsions provide excellent disinfection when used neat or diluted, e.g. at 1:100, 1:25, 1:50 dilution levels.

What is claimed is:

1. A microemulsion suitable for disinfecting a surface, comprising a surfactant, an aqueous phase comprising a bleach, and droplets dispersed in said aqueous phase, said droplets comprising an essential oil or an active thereof, and said droplets having a particle size of less than 100 nanometers.
2. A microemulsion according to claim 1 wherein said bleach is a peroxygen bleach, preferably hydrogen peroxide and/or a water soluble source thereof selected from the group consisting of percarbonates, persilicates, persulphates, perborates, peroxyacids, dialkylperoxides, diacylperoxides, preformed percarboxylic acids, organic and inorganic peroxides, organic and inorganic hydroperoxides and mixtures thereof, more preferably hydrogen peroxide.
3. A microemulsion according to any of the preceding claims which comprises from 0.01% to 15% by weight of the total microemulsion of said bleach or mixtures thereof, preferably from 0.1% to 10% and more preferably from 0.2% to 5%.
4. A microemulsion according to any of the preceding claims, wherein said essential oil is selected from the group consisting of thyme oil, lemongrass oil, citrus oil, lemon oil, orange oil, anise oil, clove oil, aniseed oil, cinnamon oil, geranium oil, rose oil, lavender oil, citronella oil, eucalyptus oil, peppermint oil, camphor oil, sandalwood oil, cedar oil, and mixtures thereof, and/or said active of essential oil is selected from the group consisting of thymol, eugenol, menthol, carvacrol, verbenone, eucalyptol, cedrol, anethol, pinocarvone, geraniol, hinokitiol, berberine, terpineol, limonene and mixtures thereof.
5. A microemulsion according to any of the preceding claims which comprises from 0.005% to 5% by weight of the total microemulsion of said essential oil or actives thereof, or mixtures thereof, preferably from 0.006% to 3%, and more preferably from 0.05% to 1%.

6. A microemulsion according to any of the preceding claims which comprises from 0.01% to 40% by weight of the total microemulsion of a surfactant or mixtures thereof, preferably from 0.05% to 15%, and more preferably from 0.1% to 12%.
7. A microemulsion according to any of the preceding claims wherein said surfactant is:

- a zwitterionic surfactant or mixtures thereof, preferably a betaine or sulphobetaine surfactant, or derivatives thereof, or mixtures thereof according to the following formula



wherein R1 is a hydrocarbon chain containing from 1 to 24 carbon atoms, preferably from 8 to 18, more preferably from 12 to 14, wherein R2 and R3 are hydrocarbon chains comprising from 1 to 3 carbon atoms, preferably 1 carbon atom, wherein n is an integer from 1 to 10, preferably from 1 to 6, and more preferably is 1, Y is selected from the group consisting of carboxyl and sulfonyl radicals and wherein the sum of R1, R2 and R3 hydrocarbon chains is from about 14 to about 24 carbon atoms, and/or

- an amphoteric surfactant or mixtures thereof, preferably an amine oxide having the following formula  $\text{R}_1\text{R}_2\text{R}_3\text{NO}$  wherein each of R1, R2 and R3 is independently a saturated substituted or unsubstituted, linear or branched hydrocarbon chain comprising from 1 to 30 carbon atoms, preferably R1 is a saturated substituted or unsubstituted, linear or branched hydrocarbon chain of from 6 to 20 carbon atoms, preferably from 8 to 16 carbon atoms, more preferably from 8 to 12, and R2 and R3 are independently substituted or unsubstituted, linear or branched hydrocarbon chains of from 1 to 4 carbon atoms, preferably of from 1 to 3 carbon atoms, and more preferably are methyl groups, or mixtures thereof.

8. A microemulsion according to any of the preceding claims which further comprises a hydroxylated solvent or mixtures thereof, up to a level of 15% by weight of the total microemulsion, preferably from 0.2% to 12% and more preferably from 0.5% to 10%.
9. A microemulsion according to claim 8 wherein said hydroxylated solvent is a glycol ether or a derivative thereof, a polyol, an alkoxyated aliphatic or aromatic alcohol, an aliphatic or aromatic alcohol, a glycol or mixtures thereof, preferably is ethylene glycol butyl ether, propylene glycol butyl ether, ethylene glycol ethyl ether, 2-(Hexyloxy)ethanol, 1,6-hexanediol, n-butoxypropoxypropanol, butyl triglycol ether, diethylene glycol butyl ether, 2-ethyl-1,3-hexanediol, 2,2,4-trimethyl-1,3-pentanediol, methyl-2,4 pentanediol, 1,2-hexanediol, 1-methoxy-11-dodecanol methoxy propanol, ethoxy propanol, propoxy propanol, decanol, ethanol, 2-propanol, benzyl alcohol, dodecaneglycol, propanediol, 2-(2-alkoxyethoxy)ethanol and/or 2-(2-alkoxyethoxy)propanol and/or 2-(2-alkoxyethoxy)butanol wherein the alkoxy group is ethyl, propyl, butyl and/or tert-butyl, and more preferably is ethylene glycol butyl ether, propylene glycol butyl ether, diethylene glycol butyl ether, benzyl alcohol, 2-propanol, ethylene glycol ethyl ether or mixtures thereof.
10. A microemulsion according to any of the preceding claims, wherein said microemulsion further comprises at least an ingredient selected from the group consisting of chelating agents, radical scavengers, thickeners, other solvents, other antimicrobial/antibacterial compounds, builders, stabilisers, bleach activators, soil suspenders, dye transfer agents, brighteners, anti dusting agents, enzymes, dispersant, dye transfer inhibitors, pigments, perfumes, dyes and mixtures thereof.
11. A microemulsion according to any of the preceding claims packaged in a spray dispenser.
12. A wipe incorporating a microemulsion according to any of the preceding claims 1 to 10.

13. The use of a microemulsion comprising a surfactant, an aqueous phase, and droplets dispersed in said aqueous phase, said droplets comprising an essential oil or an active thereof, and said droplets having a particle size of less than 100 nanometers, for disinfecting a surface.
14. The use according to the claim 13 wherein said aqueous phase further comprises a bleach, preferably a peroxygen bleach.

## INTERNATIONAL SEARCH REPORT

International application No.

PCT/US97/20512

## A. CLASSIFICATION OF SUBJECT MATTER

IPC(6) :C11D 17/50

US CL :510/417, 375, 367, 372.

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 510/417, 375, 367, 372.

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched  
NONE

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

APS: PEROXYGEN; MICROEMULSION; LIMONENE, TERPINEOL, CITRUS OIL#

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y,P	US 5,665, 268 A (DEGUERTECHIN ET AL) 09 September 1997 (09-09-97), abstract, column 5, lines 15-37; column 7, lines 60-column 8, line13; column 12, lines 24-26.	1-2, 13-14
Y,P	US 5,643,861 A (DEGUERTECHIN ET AL) 01 July 1997 (01-07-97), column 6, lines 5-25; column 9, lines 10-24; see example 1	1-3, 13-14
Y	US 4,472,291 A (ROSANO) 18 September 1984 (18-9-84), abstract, column 3, lines 18-68; column 4, lines 59-66 and claims.	1-3, 13-14



Further documents are listed in the continuation of Box C.



See patent family annex.

\*

Special categories of cited documents:

\*A\*

document defining the general state of the art which is not considered to be of particular relevance

\*E\*

earlier document published on or after the international filing date

\*L\*

document which may throw doubt on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

\*O\*

document referring to an oral disclosure, use, exhibition or other means

\*P\*

document published prior to the international filing date but later than the priority date claimed

\*T\*

later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

\*X\*

document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

\*Y\*

document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

\*A\*

document member of the same patent family

Date of the actual completion of the international search

31 DECEMBER 1997

Date of mailing of the international search report

28 JAN 1998

Name and mailing address of the ISA/US  
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# INTERNATIONAL SEARCH REPORT

International application No.  
PCT/US97/20512

## Box I Observations where certain claims were found unsearchable (Continuation of item 1 of first sheet)

This international report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:  
because they relate to subject matter not required to be searched by this Authority, namely:
2. ☐ Claims Nos.:  
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:
3. ☒ Claims Nos.: 3-12  
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

## Box II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

1. ☐ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.
2. ☐ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:
4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

- ☐ The additional search fees were accompanied by the applicant's protest.  
☐ No protest accompanied the payment of additional search fees.

## INTERNATIONAL SEARCH REPORT

International Application No.

PCT/US 99/26588

## A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 C11D3/395 C11D3/20 C11D3/34 C11D3/16 C11D3/37

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 C11D

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP 0 668 345 A (PROCTER & GAMBLE) 23 August 1995 (1995-08-23) page 4, line 3 - line 45 page 5, line 25 - line 27 claims 1-13	1,3-9
A	US 5 510 047 A (GABRIEL STEVEN M ET AL) 23 April 1996 (1996-04-23) column 6, line 49 -column 8, line 48 column 15, line 12 -column 16, line 27 claims 1-8	1,3,6-9
A	US 5 384 061 A (WISE RODNEY M) 24 January 1995 (1995-01-24) column 5, line 55 -column 6, line 16 column 9, line 37 - line 52 claim 1	1,3,6-9
	-/-	

☒ Further documents are listed in the continuation of box C.☒ Patent family members are listed in annex.

## \* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"Z" document member of the same patent family

Date of the actual completion of the international search

20 March 2000

Date of mailing of the international search report

31/03/2000

Name and mailing address of the ISA

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Authorized officer

Richards, M



# INTERNATIONAL SEARCH REPORT

International Application No

PCT/US 99/26588

## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	GB 2 279 963 A (PROCTER & GAMBLE) 18 January 1995 (1995-01-18) page 14, line 1 - line 14 claims 1-8	1,3,6-9

# INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/US 99/26588

Patent document cited in search report		Publication date	Patent family member(s)	Publication date
EP 0668345	A	23-08-1995	JP 7310098 A	28-11-1995
			AT 178938 T	15-04-1999
			AU 701112 B	21-01-1999
			AU 1171895 A	29-05-1995
			BR 9408034 A	17-12-1996
			CA 2173434 A	18-05-1995
			CN 1134735 A	30-10-1996
			CZ 9601328 A	11-12-1996
			DE 69417844 D	20-05-1999
			DE 69417844 T	18-11-1999
			EG 20629 A	31-07-1999
			EP 0653483 A	17-05-1995
			EP 0892042 A	20-01-1999
			ES 2131661 T	01-08-1999
			FI 962009 A	10-05-1996
			GR 3030580 T	29-10-1999
			HU 74708 A	28-02-1997
			JP 9510945 T	04-11-1997
			NO 961905 A	10-05-1996
			NZ 276888 A	25-03-1998
			PL 314295 A	02-09-1996
			SK 60296 A	07-05-1997
			WO 9513417 A	18-05-1995
			US 5843190 A	01-12-1998
US 5510047	A	23-04-1996	AU 3970393 A	18-11-1993
			BR 9306242 A	23-06-1998
			CA 2133445 A, C	28-10-1993
			EP 0636169 A	01-02-1995
			JP 8504220 T	07-05-1996
			WO 9321298 A	28-10-1993
US 5384061	A	24-01-1995	NONE	
GB 2279963	A	18-01-1995	CA 2126383 A, C	16-01-1995

PCT

REC'D 17 NOV 2000

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INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference CM1948M/VB		<b>FOR FURTHER ACTION</b> See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
International application No. PCT/US99/26588	International filing date (day/month/year) 10/11/1999	Priority date (day/month/year) 10/11/1998	
International Patent Classification (IPC) or national classification and IPC C11D3/395			
Applicant THE PROCTER & GAMBLE COMPANY et al.			

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.



2. This REPORT consists of a total of 6 sheets, including this cover sheet.

- ☐ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

These annexes consist of a total of sheets.

3. This report contains indications relating to the following items:

- I ☒ Basis of the report
- II ☐ Priority
- III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- IV ☒ Lack of unity of invention
- V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability: citations and explanations supporting such statement
- VI ☐ Certain documents cited
- VII ☐ Certain defects in the international application
- VIII ☒ Certain observations on the international application

Date of submission of the demand  18.05.2000	Date of completion of this report  15.11.2000
Name and mailing address of the international preliminary examining authority   European Patent Office D-80298 Munich Tel +49 89 2399 - 0 Tx 523656 epmu d Fax +49 89 2399 - 4465	Authorized officer  Hillebrecht, D  Telephone No +49 89 2399 8168  

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT**

International application No. PCT/US99/26588

**I. Basis of the report**

1. This report has been drawn on the basis of *(substitute sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to the report since they do not contain amendments (Rules 70.16 and 70.17).):*

**Description, pages:**

1-36 as originally filed

**Claims, No.:**

1-9 as originally filed

2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language: , which is:

- ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).
- ☐ the language of publication of the international application (under Rule 48.3(b)).
- ☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- ☐ contained in the international application in written form.
- ☐ filed together with the international application in computer readable form.
- ☐ furnished subsequently to this Authority in written form.
- ☐ furnished subsequently to this Authority in computer readable form.
- ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
- ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

- ☐ the description. pages:
- ☐ the claims. Nos.:
- ☐ the drawings. sheets:

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT**

International application No. PCT/US99/26588

*(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)*

6. Additional observations, if necessary:

**IV. Lack of unity of invention**

1. In response to the invitation to restrict or pay additional fees the applicant has:

- ☐ restricted the claims.
- ☐ paid additional fees.
- ☐ paid additional fees under protest.
- ☐ neither restricted nor paid additional fees.

2. ☒ This Authority found that the requirement of unity of invention is not complied and chose, according to Rule 68.1, not to invite the applicant to restrict or pay additional fees.

3. This Authority considers that the requirement of unity of invention in accordance with Rules 13.1, 13.2 and 13.3 is

- ☐ complied with.
- ☒ not complied with for the following reasons:  
**see separate sheet**

4. Consequently, the following parts of the international application were the subject of international preliminary examination in establishing this report:

- ☒ all parts.
- ☐ the parts relating to claims Nos. .

**V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement**

1. Statement

Novelty (N)	Yes:	Claims	2,5
	No:	Claims	1,3,4,6-9
Inventive step (IS)	Yes:	Claims	
	No:	Claims	1-9
Industrial applicability (IA)	Yes:	Claims	1-9
	No:	Claims	

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT**

International application No. PCT/US99/26588

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2. Citations and explanations  
**see separate sheet**

**VIII. Certain observations on the international application**

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:  
**see separate sheet**

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT - SEPARATE SHEET**

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International application No. PCT/US99/26588

Reference is made to the following documents:

- D1: EP-A-0 668 345 (PROCTER & GAMBLE) 23 August 1995
- D2: US-A-5 384 061 (WISE RODNEY M) 24 January 1995
- D3: GB-A-2 279 963 (PROCTER & GAMBLE) 18 January 1995

IV. The present application does not comply with Rule 13 PCT. Present claim 1 claims a liquid cleaning composition comprising an oxidizing agent and a radical scavenger selected from a compound of formulae (i) to (iv). However, respective compositions are already known from D1 to D3. Any of these documents discloses bleaching compositions comprising a radical scavenger.

In claim 1 of D1 are naphthoic acid, salicylic acid and methylnaphthalene sulfonate explicitly mentioned.

D2 and D3 disclose stable thickened bleaching compositions comprising besides the bleaching agent a free radical scavenging rheology stabilizing agent, i.a. naphthalene sulfonates. (Cf. D2, claim 1 and column 5, line 55 to column 6, line 46; and D3, claim 1)

Moreover, these documents teach also the use of the compounds as radical scavengers and are thus relevant for present claim 9, too.

Since the common linking concept of claims 1 and 9 is already known from D1 to D3, any compound (group of compounds) must be regarded as a single invention. However, at present it is not clear how many individual inventions are covered by present claims 1 and 9.

V. The present application does not comply with Article 33(1) and (2) PCT since the subject-matter of claims 1 and 9 does not appear to be novel in view of the cited prior art. For the reason, cf. to item IV. above.

The subject-matter of dependent claims 3, 4, and 6 to 8 is also known from D1, D2, and/or D3.

The subject-matter of claims 2 and 5 appears to be obvious in view of the cited documents, in contrast to Article 33(1) and (3) PCT.

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT - SEPARATE SHEET**

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Moreover, it is believed that a variety of compounds defined in claims 1 and 9 do not act as radical scavengers. Consequently, the present application appears to lack an inventive step, since it does not work in the entire claimed scope.

VIII.

1. Claims 1 and 9 lack clarity in contrast to Article 6 PCT, since the letters S, P, N, O, and I are usually used for the respective atoms rather than for substituents. Moreover, in view of formula (iii) it is not clear whether the naphtalene nucleus must be substituted with any of S,A,B,C,D,E,F, or G (which would e.g. include naphthalene itself) or must the nucleus be substituted by a combination of S,A,B,C,D,E,F, or G, which would require that naphtalene ring carries at most 3 hydrogen atoms.
2. The term TINOPAL and OPTIBLANC employed in claim 5 and appearing to be registered trade marks have no precise meaning as they are not internationally accepted as standard descriptive terms, thereby rendering the definition of the subject-matter of this claim unclear (Article 6 PCT).
3. Terms like "poly hydroxy styrene" used in claim 2 lack clarity since they are ambiguous. They may stand for a styrene monomer carrying a plurality of hydroxy groups as well as for polymers obtainable from monomeric hydroxy styrene. (Article 6 PCT)
4. Terms like "oxidizing agent of the invention" used on pages 6, 7, and 10 which are not directed to a composition of claim 1 or a respective use, cause lack of clarity when used to interpret the claims (Article 6 PCT).
5. The objections raised under items 1. and 3. apply also to the respective parts of the specification.



# TENT COOPERATION TREATY

## PCT

### INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference <b>CM1948M/VB</b>	<b>FOR FURTHER ACTION</b> <small>see Notification of Transmittal of International Search Report (Form PCT/ISA/220) as well as, where applicable, Item 5 below.</small>	
International application No. <b>PCT/US 99/ 26588</b>	International filing date (day/month/year) <b>10/11/1999</b>	(Earliest) Priority Date (day/month/year) <b>10/11/1998</b>
Applicant  <b>THE PROCTER &amp; GAMBLE COMPANY et al.</b>		

This International Search Report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This International Search Report consists of a total of 3 sheets.

☒ It is also accompanied by a copy of each prior art document cited in this report.

**1. Basis of the report**

a. With regard to the language, the international search was carried out on the basis of the international application in the language in which it was filed, unless otherwise indicated under this item.

☐ the international search was carried out on the basis of a translation of the international application furnished to this Authority (Rule 23.1(b)).

b. With regard to any nucleotide and/or amino acid sequence disclosed in the international application, the international search was carried out on the basis of the sequence listing:

☐ contained in the international application in written form.

☐ filed together with the international application in computer readable form.

☐ furnished subsequently to this Authority in written form.

☐ furnished subsequently to this Authority in computer readable form.

☐ the statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.

☐ the statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished

2. ☐ Certain claims were found unsearchable (See Box I).

3. ☐ Unity of invention is lacking (see Box II).

4. With regard to the title,

☒ the text is approved as submitted by the applicant.

☐ the text has been established by this Authority to read as follows:

5. With regard to the abstract,

☒ the text is approved as submitted by the applicant.

☐ the text has been established, according to Rule 38.2(b), by this Authority as it appears in Box III. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority.

6. The figure of the drawings to be published with the abstract is Figure No. \_\_\_\_\_

☐ as suggested by the applicant.

☐ because the applicant failed to suggest a figure.

☐ because this figure better characterizes the invention.

☐ None of the figures.

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A CLASSIFICATION OF SUBJECT MATTER  
IPC 7 C11D3/395 C11D3/20 C11D3/34 C11D3/16 C11D3/37

**B. FIELDS SEARCHED**

IPC 7 C11D

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

### C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP 0 668 345 A (PROCTER & GAMBLE) 23 August 1995 (1995-08-23) page 4, line 3 - line 45 page 5, line 25 - line 27 claims 1-13	1,3-9
A	US 5 510 047 A (GABRIEL STEVEN M ET AL) 23 April 1996 (1996-04-23) column 6, line 49 -column 8, line 48 column 15, line 12 -column 16, line 27 claims 1-8	1,3,6-9
A	US 5 384 061 A (WISE RODNEY M) 24 January 1995 (1995-01-24) column 5, line 55 -column 6, line 16 column 9, line 37 - line 52 claim 1	1,3,6-9

-/-

**X** Further documents are listed in the continuation of box C.

**X** Patent family members are listed in annex.

\* Special categories of cited documents :

\*P\* document published prior to the international filing date but later than the priority date claimed

"&" document member of the same patent family

**20 March 2000**

31/03/2000

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**Authorized officer**

**Richards, M**

# INTERNATIONAL SEARCH REPORT

International Application No.  
PCT/US 99/26588

## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	<p>GB 2 279 963 A (PROCTER &amp; GAMBLE)  18 January 1995 (1995-01-18)  page 14, line 1 - line 14  claims 1-8</p>	1,3,6-9

# INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/US 99/26588

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